EXPLICIT APPLICATION OF SWANEPOEL'S METHOD FOR THE ANALYSIS OF Sb_2O_3 FILMS

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Abstract. *Swanepoel's method can be applied successfully for the determination of essential optical parameters for films exhibiting interference pattern-type of optical transmission spectra, such those made of antimony trioxide deposited on glass. Refractive index dispersion, absorption spectrum, the real and the imaginary parts of the dielectric constant and the optical conductivity are some of the important features that can be determined by using this valuable method. Also, calculation of films' thicknesses can be inferred by using Swanepoel's method. Still, care has to be taken concerning the estimation of film's thickness from pairs of transmission maxima and/or minima, since important variations may occurs from several computations based on the same spectrum. This article makes a detailed presentation of the application of the mentioned method for the determination of some of the optical features of vacuum-deposited* Sb_2O_3 *films.*

Keywords: *film, optical transmittance, Swanepoel's method, refractive index, thickness.*

1. INTRODUCTION

Antimony trioxide (Sb_2O_3) is a chalcogenide semiconductor compound of growing interest, because of its potential use in paints and adhesives preparation, for various optoelectronic devices, such as flat panel displays, light emitting devices, solar cells or as gas sensor. These various applications are due to the particular properties of Sb_2O_3, such as large energy bandgap, high electrical conductivity and simple, cubic-type of structure at room temperature [1].

In order to design optoelectronic devices based on a specific film, it is necessary to calculate and analyze its optical constants, such as: refractive index, n, absorption coefficient, α, optical band gap, Eg, etc. [2, 3]. All these optical parameters, along with some others can be calculated solely from the transmittance spectrum of a film exhibiting successive local maxima and minima (see Fig. 1), by means of the method proposed by Swanepoel [4]. This happens if several conditions are simultaneously met: 1. if the film thickness is uniform, thus, the interference effect giving rise to the particular aspect of the transmission spectrum, with peaks and valleys**;** 2. if the film is deposited on a transparent substrate**;** 3. if the film is several orders of magnitude thinner than the substrate (generally, the substrate is 1-2 mm thick, while the film has tens-hundreds of nanometers thickness) [1, 4, 5].

The local maxima and minima in such a particular transmission spectrum can be also used to calculate the film thickness, which is a very attractive feature. This is because the film thickness is necessary in order to calculate various optical and other types of physical parameters and the methods usually employed for its calculation impl**y** scratching the film (profilometry, optical interferometry with a Linnik microscope, etc.).

It is important to study various optical parameters of a certain deposited film since they show sensitivity to numerous physical properties of the substances under study, starting from their nature, to their structure, preparation and deposition conditions [6] etc.

This article presents in details the steps followed in order to make a complete optical characterization of some antimony trioxide films, starting from their optical transmission spectrum, by applying the algorithm proposed by Swanepoel. Formulas and graphs are given for each step of the procedure.

2. EXPERIMENTAL DETAILS

Films of Sb_2O_3 were deposited by conventional thermal evaporation in vacuum at $5 \cdot 10^{-5}$ torr, onto chemically-cleaned microscope glass substrates, maintained at several different temperatures. Only one such type of Sb_2O_3 film is analyzed within this article, namely the one deposited at 20^0 C temperature of the glass substrate.

The optical transmission spectra of the films were acquired at room temperature with a Perkin Elmer Lambda 35 spectrometer, operated in air, at normal incidence, in the 190 nm – 1100 nm spectral range, with a 2 nm step. The optical transmission spectrum of the glass substrate alone was also acquired. Thus, the effective transmittance of the Sb_2O_3 films could be extracted from the total transmission factor, T of the film deposited onto glass, according to the following formula (see inset in Fig. 1) [2]:

$$T_{eff} = 100 \cdot \frac{T}{T_{glass}} \; (\%) \qquad (1)$$

where T_{eff} is the effective transmission factor of the film and T_{glass} is the transmission factor of the glass used as substrate. Each transmittance factor in formula (1) is expressed in percents (%).

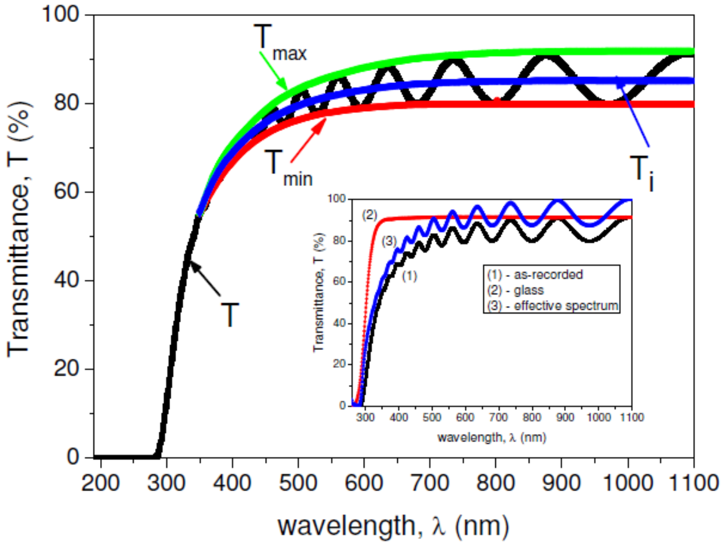

Fig. 1. Transmission spectra for the Sb_2O_3 film deposited on glass (in the inset: (1) – including the substrate; (2) – for the glass substrate alone; (3) – effective transmittance for Sb_2O_3).

3. PROCEDURE AND EXPERIMENTAL RESULTS

In the following, the refractive indexes of the film and of the substrate will be denoted by *n* and *s*, respectively and their corresponding transmittances will be known as T and T_s, respectively. Also, only the case of normal incidence of optical radiation on the studied films will be considered.

The peaks and valleys-aspect of the transmission spectra of a film for which one can apply the Swanepoel algorithm appear in the region of low and medium absorption. The first step in the procedure is to identify the position (wavelength) and height (transmittance value) of each local minimum and maximum in the transmission spectrum of the entire film+substrate structure. These values for the Sb_2O_3 film under study, extracted from Fig. 1 are presented in Table 1.

The wavelengths of the interference maxima and minima at normal incidence of the optical radiation of a certain film satisfy the following condition [7]:

$$2n \cdot d = m \cdot \lambda \quad (2)$$

where *m* is an integer number for interference maxima and semi-integer for interference minima, respectively.

Table 1. Positions and values of transmission maxima and minima

Transmission minima		Transmission maxima	
λ (nm)	**T (%)**	**λ (nm)**	**T (%)**
362	58.804	354	56.244
382	63.757	370	62.569
408	68.438	393	68.669
440	72.482	424	74.318
480	75.568	458	78.914
530	77.857	503	82.917
595	79.084	561	86.111
680	79.985	633	88.303
798	79.945	734	89.967
973	79.87	873	90.825
-	-	1100	91.193

Thus, by applying the preceding formula for any pair of consecutive interference maxima or minima, corresponding to λ_1 and λ_2 wavelengths and to n_1 and n_2 refractive indexes, the thickness, d of the film can be estimated by using the next expression [5]:

$$d = \frac{\lambda_1 \cdot \lambda_2}{2 \cdot (\lambda_1 \cdot n_2 - \lambda_2 \cdot n_1)} \quad (3)$$

Table 2 presents the data and the results for the thickness, *d* calculation of the antimony trioxide film under study from all the possible pairs of successive transmission maxima and minima, respectively. The data in italic denote resulting values much higher than the rest of the data set for film thickness.

Table 2. Computation of film thickness from pairs of transmission maxima and minima

Crt. No.	Extremum-type	λ_1 (nm)	λ_2 (nm)	n_1	n_2	d (nm)	$\bar{d}$ (nm)
1.	min	362	382	1.73395	1.80787	***8730.095***	**1527.1769**
2.	min	382	408	1.80787	1.78829	1430.287	
3.	min	408	440	1.78829	1.79538	1652.048	
4.	min	440	480	1.79538	1.81391	1658.760	
5.	min	480	530	1.81391	1.82916	1525.628	
6.	min	530	595	1.82916	1.85662	1511.142	
7.	min	595	680	1.85662	1.88675	1446.184	
8.	min	680	798	1.88675	1.91929	1353.154	
9.	min	798	973	1.91929	1.92856	1181.895	
10.	max	354	370	1.65481	1.79369	***2901.263***	
11.	max	370	393	1.79369	1.79927	1855.180	
12.	max	393	424	1.79927	1.78626	1368.297	
13.	max	424	458	1.78626	1.80466	1834.380	
14.	max	458	503	1.80466	1.8224	1576.074	
15.	max	503	561	1.8224	1.84045	1460.271	
16.	max	561	633	1.84045	1.86986	1530.483	
17.	max	633	734	1.86986	1.90088	1372.833	
18.	max	734	873	1.90088	1.92387	1295.306	

Thus, if one should use only one pair of transmission factor extremes for the computation of the thickness, d, this would lead to imprecise estimation of this parameter. The average value of all the possible calculations should be considered as closest to the true value of d. Still, the values of d too far from most of the other results should be excluded from the average. Thus, in table 1, the average value of d, denoted as $\overline{d}$, does not include the extremely high values of d in italic. The average value of d including the 2 extreme values in italic is: $\overline{d} = 2098.9772$ nm.

Because of the reasons stated above, in the following computations the average thickness of the Sb_2O_3 film will be taken as: $\overline{d} = 1527.1769$ nm.

The complex refractive index of a homogeneous film, with uniform thickness is defined by:

$$\tilde{n} = n - i \cdot k \tag{4}$$

where n and k represent the real part (the proper refractive index) and the imaginary part (the extinction coefficient or the absorption index), respectively.

According to the algorithm proposed by Swanepoel, in order to determine the refractive index of the film under study, one has to build the maximum and minimum transmittance envelope **plots** T_M (λ) and T_m (λ), respectively (see Fig. 1). With a proper computer program, the interpolation of these envelopes can be done, in order to have a more accurate determination of the optical constants.

The next step is to calculate the average transmittance, T_i of the film, according to:

$$T_i = \frac{2 \cdot T_M \cdot T_m}{T_M + T_m} \tag{5}$$

The resulting $T_i = T_i(\lambda)$ for the studied film is also presented in Fig. 1.

The optical transmission spectra proved that the film is highly transparent (transmittance around 75–90%) at wavelengths higher than 400 nm. The appearance of interferences fringes is an indication of the thickness uniformity of the films. The sharp fall of transmittance at the band edge indicates the crystallinity of the films [2].

By using the maxima and minima envelopes, $T_M = T_M(\lambda)$ and $T_m = T_m(\lambda)$, respectively, the refractive index $n = n(\lambda)$ can be obtained as explained below.

First, the refractive index of the substrate (microscope glass slide – in this case), *s*, has to be calculated from the transmittance data, T_s of the substrate alone, according to the next formula:

$$s = \frac{1}{T_s} + \left(\frac{1}{T_s^2} - 1\right)^{1/2} \tag{6}$$

Then, the refractive index, *n*, of the film is computed as:

$$n = \left[N + \left(N^2 - s^2\right)^{1/2}\right]^{1/2} \tag{7}$$

where N is a parameter given by [8]: $N = 2 \cdot s \cdot \frac{T_M - T_m}{T_M \cdot T_m} + \frac{s^2 + 1}{2}$ (8)

Fig. 2 presents the dispersion of the refractive index for the specified Sb_2O_3 film corresponding to low and medium absorption regions, i. e. from the part of the transmission spectrum with interference maxima and minima.

The values of the refractive index into the strong absorption region can be obtained with rather good precision by extrapolating the $n = n(\lambda)$ plot from the weak and medium absorption regions. It can be notice from Fig. 2 that the refractive index of the Sb_2O_3 film varies between 1.6127 at 350 nm and 1.9280 at 1100 nm, having a local prominent maximum of 1.8110 at 379 nm.

In order to find the absorption coefficient, α of the film in the high transmittance region, one has to compute the following parameter:

$$x = e^{-\alpha \cdot d} = \frac{F - [F^2 - (n^2 - 1)^3 \cdot (n^2 - s^4)]^{1/2}}{(n-1)^3 \cdot (n - s^2)} \tag{9}$$

where F is given by:

$$F = \frac{8n^2 \cdot s}{T_i} \tag{10}$$

Thus, the absorption coefficient α can be calculated as: $\alpha = -\frac{1}{d} \cdot \ell n x$ (11)

The resulting absorption spectrum for the film under study is given in Fig. 3, together with an exponential-type of fit of the plot (see fitting formula in the inset).

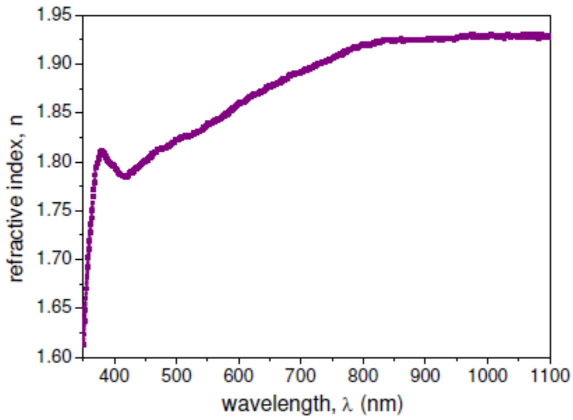

Fig. 2. Dispersion of the refractive index for the Sb_2O_3 film deposited on glass.

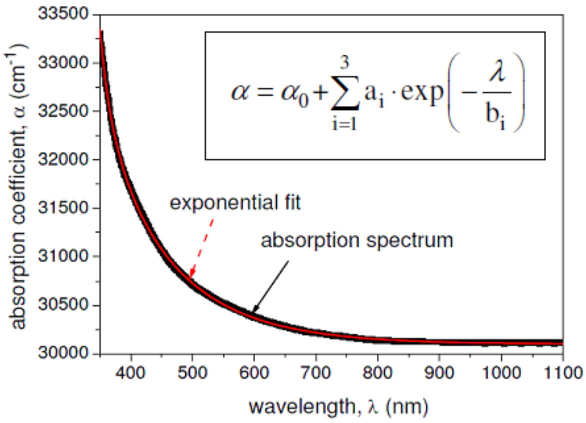

Fig. 3. Absorption spectrum for the analyzed film, with multiple exponential decay fit.

The absorption index or the extinction coefficient, k, can be calculate from the next expression:

$$k = \frac{\alpha \cdot \lambda}{4\pi} \tag{12}$$

Fig. 4 presents the plot of the extinction coefficient, *k* versus wavelength, *λ*, together with a linear fit of the plot (see formula in the inset).

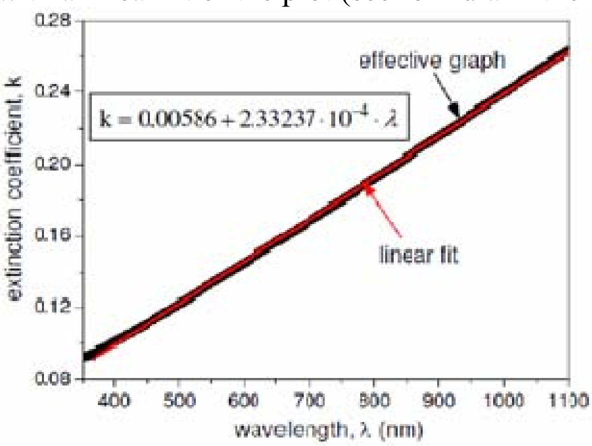

Fig. 4 Extinction coefficient versus wavelength, with linear fit of the plot.

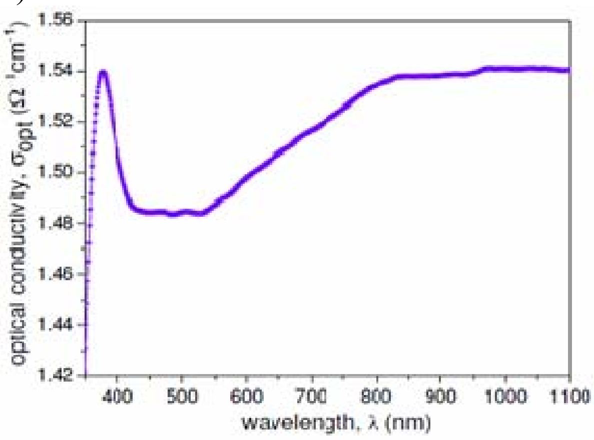

Fig. 5 The optical conductivity versus wavelength for the analyzed Sb_2O_3 film.

The optical conductivity can be computed according to [8]: $\sigma = \frac{\alpha \cdot n \cdot c}{4\pi}$ (13)

The following transformation has to be made from the CGS system to the MKS system, in order to express correctly the optical conductivity [11]:

$$\sigma(\text{in } \Omega^{-1}\cdot\text{cm}^{-1}) = \frac{\sigma(\text{in C.G.S units})}{9\cdot 10^{11}} \tag{14}$$

The resulting plot of the optical conductivity against wavelength is given in Fig. 5.

It can be noticed that the optical conductivity of the Sb_2O_3 film deposited on glass substrate at 20^0 C, by classic vacuum thermal evaporation has values around 1.48 $\Omega^{-1}\cdot\text{cm}^{-1}$, in the range of the electrical conductivities for semiconductors, i. e. between $10^{-8}\,\Omega^{-1}\cdot\text{cm}^{-1}$ and $10^3\,\Omega^{-1}\cdot\text{cm}^{-1}$.

4. CONCLUSIONS

Starting only from the transmission spectrum of a transparent film, exhibiting successive local maxima and minima, the method proposed by Swanepoel allows for the determination of essential optical parameters for the studied films. Also, the film thickness can be computed according to the same method. This algorithm was applied for a film of antimony trioxide deposited on glass by thermal evaporation in vacuum. Several optical parameters were deduced.

Among them, the refractive index resulted to reach values above 1.6 for the 350-1100 nm spectral range. The optical conductivity for the same film has values in the range of unity, expressed in $\Omega^{-1}\cdot\text{cm}^{-1}$. Numerical fit was possible for the absorption spectrum against wavelength and also for the extinction coefficient against the same variable. Furthermore, a 2.093 eV energy optical bandgap was estimated (not showed here), belonging to the allowed direct-type of electronic transition of the studied film.

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